《報文》 HWAHAK KONGHAK Vol. 19, No. 4, August 1981, pp. 291—302
(Journal of the Korean Institute of Chemical Engineers)

# 圓環流動層에서의 氣體混合特性

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(접수 1981. 8. 10)

# Gas Mixing in an Annular Fluidized Bed

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(Received August 10, 1981)

## 요 약

Annular 형태의 유동층에서 기체의 축 방향 혼합계수와 측면방향(lateral) 혼합계수를 구하였다. Annular 단면에서 기체유속, 입자크기 및 유동화입자의 유동화조건이 다른 입자의 첨가가 기체의 혼합에 미치는 영향을 결정하였다.

측면 방향혼합계수의 산출은 diffusion type model을 제안하여 적용하였으며 축 방향혼합에서는 Schügerl의 Model를 사용하였다. 측면방향 기체혼합계수와 축 방향혼합계수 모두 기체의 유속이 증가함에 따라 지수함수적으로 증가하였고 입자크기의 증가에 따라 측면방향 혼합은 감소하며 기체 유속의 영향은 축 방향혼합계수가 측면방향보다 크게 나타났다.

이들 기체혼합계수는 본 실험의 조건에서 입자크기와 기체유속의 함수로서 잘 표현되었다.

## ABSTRACT

Lateral and axial gas phase mixing have been measured in the annular section of an annular fluidized bed which was constructed from two pieces of 7.6 cm-diameter and 2.54 cm-diameter plexiglass cylinder.

Effects of gas velocity, particle size and weight percent of added inert particles on gas

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phase mixing have been determined. For lateral mixing, a diffusion type model was proposed to analyze the present system and Schugerl model H was employed to calculate the axial gas mixing coefficients.

Both gas mixing coefficients increased exponentially with gas flow rate. Lateral gas mixing decreased with particle size. The addition of inert particles increased lateral gas mixing however, the reverse trend was observed for axial gas mixing with the addition of inert particles.

Axial and lateral gas mixing coefficients were correlated in terms of gas flow rate and particle size in an annular fluidized bed.

## INTRODUCTION

In gas-solid fludized bed reactors, the intensity of gas mixing which is closely related to the mixing of solids is one of the most important factors to influence the selectivity yield, and chemical reaction conversion especially in complicated parallel and series reactions. There, gas and solid mixing, CTD (contact time distribution) and RTD(residence time distribution) have been studied extensively.1~6) Three tracer techniques have been employed namely, unsteady state injection of tracer in one dimension, continuous injection at a point, and continuous injection at a plane in the fluidized bed. Annular fluidized bed reactors would be an attractive configuration where uniform temperature gradient and high heat transfer rate are important, as in the two chemical reactions take place simultaneously. For instance, one reaction is exothermic and the other is endothermic, it is desirable to take advantage of heat recovery from exothermic reaction zone to endothermic zone by means of annular shape reactor. Although many investigations have been reported on gas mixing for gas flow through fluidized beds of circular section, no experimental work is available in the literature with regard to gas mixing characteristcs of an annular fluidized bed. Experimental data on pressure drop in fixed and expanded beds of annular section have been reported by Kasthuri and Laddha.7) An annular type fluidized bed for solids feeding device has been developed by Chin et al.8)

When two sets of closely graded particles differing only in average size are mixed and fluidized by a gas, the resulting mixture has a minimum fluidizing velocity, $v_{mf}$, intermediate between those of the two components. Rowe and Nienow9) determined the $v_{mf}$ of multicomponent particle mixtures, Cheung et al.10) and Wen and Yu11) have been measured $v_{mf}$ variation with the change of fluidization conditions. The total pressure drop, pressure drop profiles, bed expansion and bed porosity for a variety of binary mixtures over a wide range of gas velocities have been studied by Chiba et al.12) in a circular fluidized bed.

In this study, the addition of inert particles in the bed of solids on gas phase mixing in an annular fluidized bed are also reported.

## Analysis of Lateral Dispersion

Gas mixing in the lateral direption is unique in annular fluidized bed, and these mixing phenomena in the annular section

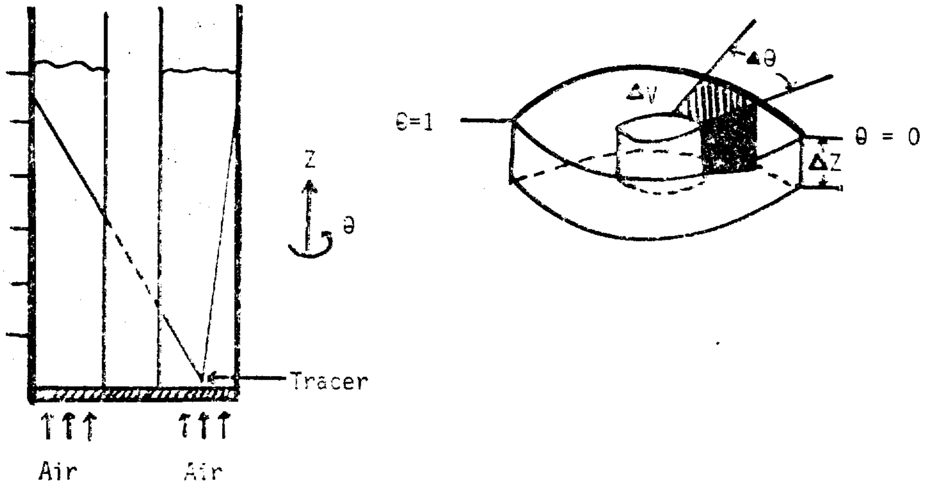
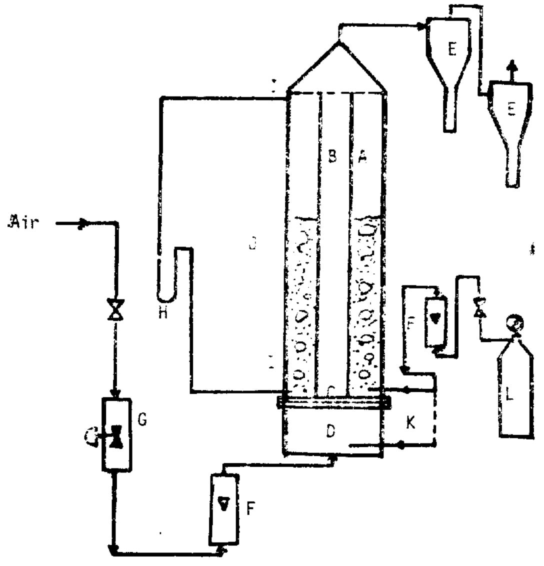


**Fig.** 1. Schematic Diagram of Lateral Mixing Model



A : Outer Bed
B : Inner Bed
C : Distributor
D : Air Box
E : Cyclone
F : Rotameter
G : Regulator
H : Manometer
I : Pressure Tap
J : Sampling Tap
K : Tracer Injection
L : $CO_2$-cylinder

**Fig.** 2. Apparatus

may be explained by the tracer movement.

A schematic diagram for lateral dispersion shown in *Fig. 1* which can be used to facilitate the understanding of tracer movement. Tracer injected into the annular section is mixed axially and laterally by means of air-bubble flow and thus if the tracer is injected such that considerable concentration gradient is established according to the axial direction as in the case of plane source, then gas mixing in both direction should be considered. However, tracer is injected into one point of the one side of the annular section and it is sampled in opposite side, axial dispersion may be neglected with respect to lateral dispersion. Except for the position of tracer injection, the direction of concentration gradient is alternating with axial coordinate until about half of the lateral direction and beyond the half, the concentration gradient is against the convective flow direction which is large enough to neglect the effect of axial dispersion.

Thus, to analyze the mixing mechanism in lateral direction in the annular section, a simple model is introduced with assumptions of

(1) end effect of the bed is negligible,

(2) steady-state condition is assumed, and this condition can be achieved by step injection of tracer,

(3) concentration profile in the radial direction is flat,

(4) axial dispersion effect is neglected in determination of lateral dispersion coefficient,

(5) adsorption effect is neglected by using nonporous particles(non-catalytic),

(6) the symmetry of tracer concentration gradient in lateral direction in both sides of the tracer injeciion point.

According to the above assumptions, material balance of tracer in segement dV can be modified as

$$\frac{\partial C_A}{\partial t} = D_\theta \frac{\partial^2 C_A}{\partial \theta^2} - V \frac{\partial C_A}{\partial Z} + D_z \frac{\partial^2 C_A}{\partial Z^2} \tag{1}$$

At steady-state, with the 4th assumption, equation (1) can be written as

$$V \frac{\partial C_A}{\partial Z} = D_\theta \frac{\partial^2 C_A}{\partial \theta^2} \tag{2}$$

which is the model equation to obtain lateral mixing coefficient. The boundary conditions for this equation can be set as

$$\frac{\partial C_A}{\partial \theta} = 0 \qquad \text{at } \theta = 0 \tag{3}$$

$$\frac{\partial C_A}{\partial \theta} = 0 \qquad \text{at } \theta = 1 \tag{4}$$

$$C_A = C_{A0} \qquad \text{at } Z = \infty \tag{5}$$

where $C_{A0}$ is tracer concentration in the bed at which tracer is completely mixed with fluidizing gas.

The initial concentration profile of tracer around the injection point can be obtained by exponential decay form, i.e.,

$$C_{Ai} = C \exp\left(-\theta^N\right) \tag{6}$$

in which $N$ was determined by repeating the iteration for various $N$ values until overall tracer concentration $C_{A0}$ was obtained at infinite distance $Z$, whatever dispersion coefficient and gas velocity may be employed.

The model equation was solved with the boundary conditions by means of the numerical implicit method.

## EXPERIMENTAL

### A. Apparatus

The schematic diagram of apparatus is shown in *Fig. 2*. It consists of annular column with sampling taps, pressure taps, cyclones, distributor and gauges for meaurements of air velocity, pressure drop and tracer injection rate.

Column and Distributor

To make annular fluidized bed, 2.54 cm-ID plexiglass cyclinder was concentrically mounted inside the 7.62 cm-ID plexiglass cyclinder and its equivalent diameter($D_0$-$D_i$) is 4.4 cm. The bed height is 120 cm for both columns.

The solid particles(74~250$\mu$) were supported on a ceramic sintered distributor which made of 74 $\mu$ glass particles and it served as the gas distributor.

Oil free compressor air was fed to the column through a pressure regulator, filter and calibrated rotameter. It was admitted to the annular section of the bed through a 2.54 cm pipe in the air box.

Two pressure taps were mounted flush with the wall of the column at the bottom and the top of the column height. The static pressure drop of the bed was measured with a liquid manometer.

The solids used were either 74, 105, 180, and 250 $\mu$ pyrite with density of 3.38 g/cm$^3$. The unfluidized bed height was 40 cm. The

**Table 1.** Summary of Experimental Conditions

Annular Fluidized Bed
  Inner Diameter : 2.54 cm-ID
  Outer Diameter : 7.62 cm-ID
  Bed Height : 120 cm
Air Box Height : 15 cm
Gas Velocity Range : 3-40 cm/sec
Solid Material : Pyrite(0.074-0.25 mm)
Mean Density of Pyrite : 3.38 g/cm$^3$
Minimum Fluidization Velocity : 1.02-5.25 cm/sec
Inert Particle : Glass beads (1 mm)
Weight Variation of Inert Particle : 5-15 %

added inert particles used was 1 mm glass beads with density of 2.5 g/cm$^3$.

Seven tracer sampling taps were mounted the opposite side of the tracer source at 5cm height intervals. Each tap equipped with valve and a rubber tube to connect the sampling bottle.

Details of the experimental conditions are summarized in *Table 1.*

### B. Tracer Measurement Technique

Air was introduced into the bed of solids at the desired superficial gas velocity. When steady-state was reached, tracer, $CO_2$ gas was injected into the bed by way of step function through the injection point which is located 1.5 cm above the distributor(point source).

The concentration of tracer was monitored by gas chromatography from the samples of $CO_2$-air mixtures taken at various height of sampling taps.

For axial gas mixing experiments, five seconds pulse of tracer was injected into the air box for even distribution(plane source).

Pressure drop in the bed also measured by means of a water manometer which is connected to the pressure taps.

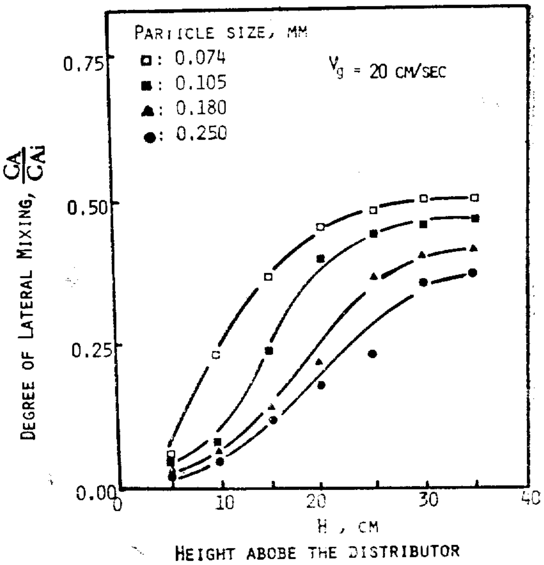


**Fig. 3.** Degree of Gas Mixing Along The Bed Height

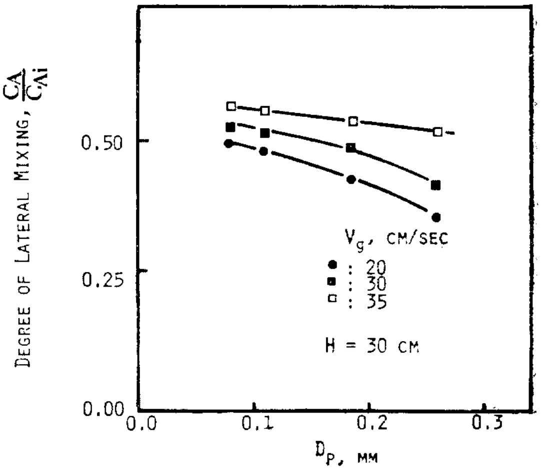


**Fig. 4.** Effect of Particle Size on Lateral Gas Mining

Same experimental procedures were also repeated in order to determined the effect of the added inert particles on gas mixing in the annular bed. The content of inert particles was varied from 5 to 15 wt. %.

## RESULTS AND DISCUSSION

Gas mixing was examined in both directions, lateral and axial, with the variations of particle size, gas velocity and contents of inert particles.

### 1. Lateral Gas Mixing

Tracer concentration profile and degree of gas mixing along the bed height illustrated the degree of gas mixing in annular fluidized bed.

The trend of gas mixing with bed height was nearly identical with different particle size(*Fig. 3*). The degree of lateral mixing decreased with increasing particle size at given gas velocity.

Effect of particle size on lateral gas mixing may be seen in *Fig. 4*. The degree of lateral gas mixing increased with gas velocity and it decreased with particle size.

The non-uniformity of gas velocity in radial direction is responsible for the spread of the hydrodynamic RTD and gas mixing in fluidized bed, and considered that RTD is also influenced by mass transfer between bubble and emulsion phases.[4,5,13] In an annular fluidized bed, gas mixing of lateral direction may be similar to that of radial direction in a fluidized bed of circular section. The gas velocity in side the bubble phase may differs from that of the rising bubble. This velocity difference may attribute to the driving force of mass transfer between the two phases. In addition, there is diffusion between the boundaries of the two phases due to the concentration gradient of bubble and dense phases. The mass transfer due to diffusion as a function of bubble size and diffusivity has been developed by Baird and Davidson[14] Mass transfer due to velocity gradient found to be increased with particle size and gas flow rate.[6]

Lateral gas mixing coefficient increased exponentially with gas velocity at given particle size(*Fig. 6*). The difference of coefficient between smaller and larger particle systems was reduced with gas velocity. It has been known that bubble size in the bed of smaller particles is larger than that of larger particles at given gas velocity. Thus velocity gradient in the bed of smaller par-

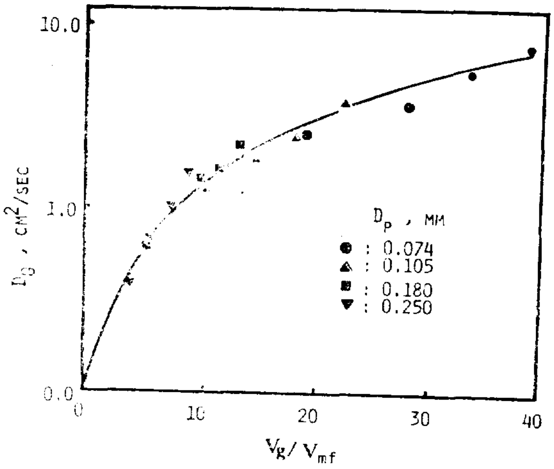


**Fig. 5.** Lateral Gas Mixing Coefficient VS $V_g/V_{mf}$

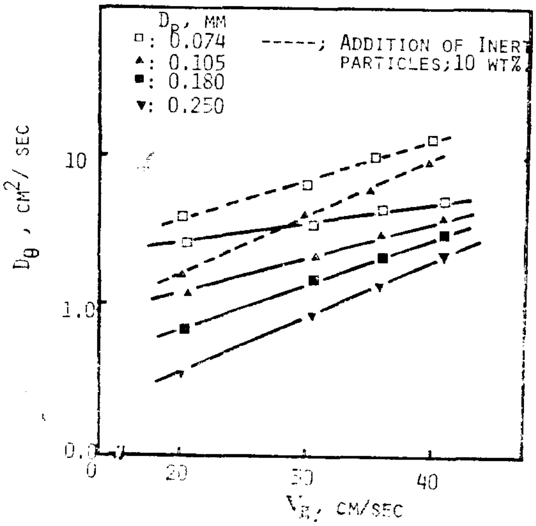


**Fig. 6.** Effect of Gas Velocity on Lateral Gas Mixing Coefficient

ticles is higher than that of larger paricle bed. However, the mass transfer coefficient due to diffusion is proportional to $d_B^{-1/4}$. That may indicate that diffusion effect on mass transfer decreases with gas velocity in the bed of smaller particles. On the basis of present findings, the effect of diffusion on mass transfer may be stronger than that of velocity gradient.

The lateral gas mixing coefficient may be expressed in terms of fluidization number $(V_g/V_{mf})$ for all particle sizes(*Fig. 5*). The lateral gas mixing coefficients increased exponentially with fluidization number since the mixing is mainly caused by gas flow rates in the beds of all particle sizes. In addition, gas mixing may be strongly caused by the bubble motions during bubble coalescence and disintegration. However, it has been well known that the bubble size may grow with gas velocity in fluidized bed.

In a fluidized bed reactor, the presence of large gas bubbles may lead to a reduction in gas-particle contact and thus to reduce process efficiency. In large reactors it is necessary to enhance the contacting of the components by increasing bubble population through the use of fixed baffles or screens[15] within the bed. However, this internals have several drawbacks such as particle attrition, higher power consumption and higher equipment cost. Therefore, the concept of mobile bubble breaker have been developed.[16,17]

With the concept of mobile bubble breaker, 1 mm glass beads as the breaker was added in the bed of pyrite particles with weight variation of 5-15%. As we may expected the rapid mixing zone lowered with the addition of inert particles as shown in *Fig. 7*.

As particle size reduced from 250 to 74 $\mu$, the degree of gas mixing was more pronounced by the addition of inert particles. With the addition of inert particles, larger bubbles may break up to smaller one, therefore, velocity gradients between inside the bubble and rising bubbles become smaller. When the gas velocity gradient and bubble size are small the contribution of diffusion between boundaries of two phases, bubble and dense phases, on mass transfer coefficient increased at the given gas velocity. Thus, degree of gas mixing increased sharply in the bed of inert particles with decreasing particle size (*Fig. 8*).

At lower gas velocities, added inert particles are almost rested in the lower part of the bed, slightly above the distributor, which is termed as "Jetsam"[12] which is acted as a part of a distributor.

At higher gas velocities, some of the added inert particles are fluidized with the particles in the bed, Floatsam.[12] From the visual observation, there are Jetsam and Jetsam-Floatsam areas appeared in the bed. These partial mixing of the solids may improve gas mixing by way of breaking bubbles. These partial mixing zone becomes broad as the increase of amount of inert particles and gas velocity(*Fig. 6*). The effect of added inert particles on lateral gas mixing increased with gas velocity and it decreased with particle size as shown in *Fig. 6*.

## 2. Axial Gas Mixing

In axial gas mixing experiments, residence time distribution measurements were made as described earlier. In order to obtain axial mixing coefficient, Schugerl Model H[3] was employed.

$$\sigma^2 = \frac{2}{Pe} + \frac{3}{Pe^2} \tag{7}$$

in which $\sigma^2$ and $Pe$ are second moment of

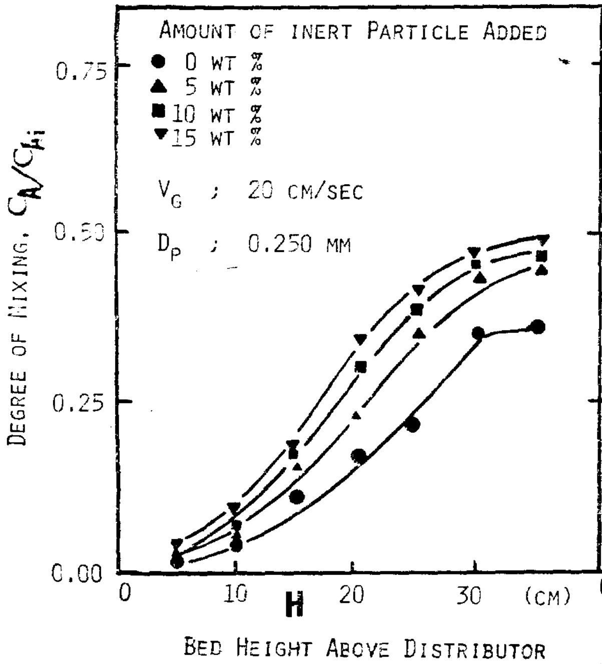


**Fig. 7.** Effect of the Inert Particles on Lateral Gas Mixing

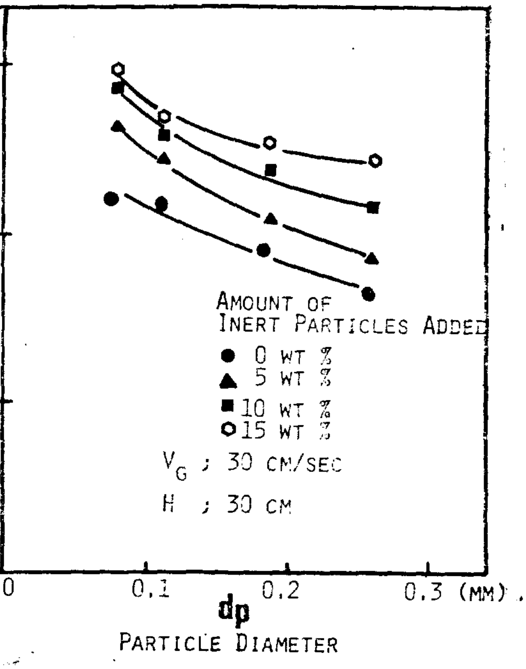


**Fig. 8.** Effect of the Inert Particles on Axial Gas Mixing

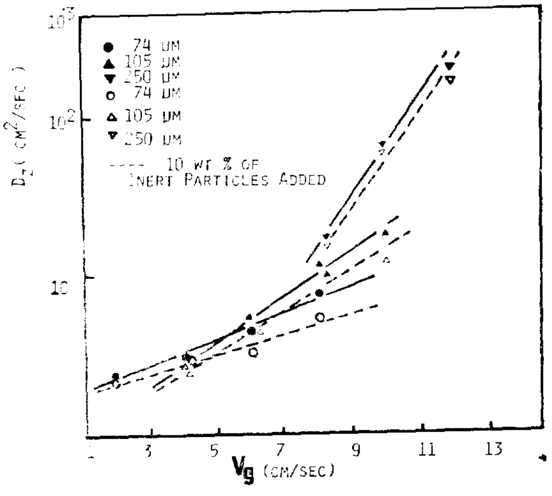


**Fig. 9.** Effect of Inert Particles on Axial Gas Mixing

tracer response curve about the mean and Peclet number, respectively.

The mechanism of gas mixing in axial direction can be interpreted as the same manner of lateral gas mixing. Bubble properties and velocity gradient between bubble and emulsion phases are the major factors to influence axial mixing in fluidized bed. However, in this case convective mass transfer due to velocity gradient may be more important than that in lateral mixing.

As may be seen in *Fig. 9*, axial mixing coefficients were much higher than those of lateral mixing. The coefficient increased exponentially with gas velocity. It may be attributed to the concentration gradeint in axial direction which may influences diffusion between bubble and dense phases directly and velocity gradient effect on mass transfer would be greater than that in lateral mixing. The convective contribution of mass transfer due to velocity gradient was found to be less than 3 % of the diffusion in the bed of 250 $\mu$ and less than 60 % in the bed of 500 $\mu$ glass beads.6) Effect of

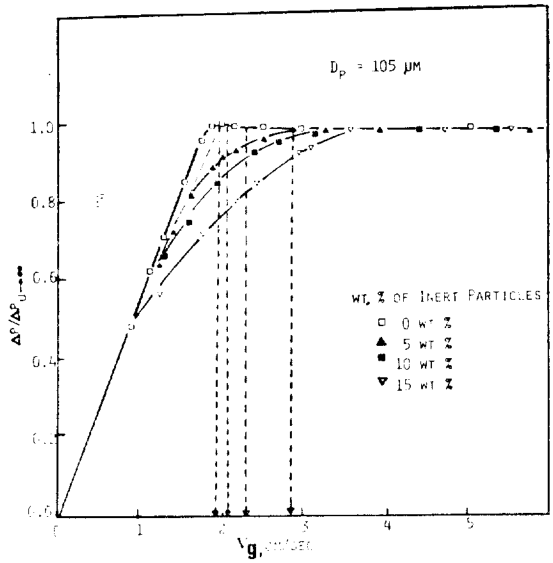


**Fig. 10.** Effect of Inert Particles on Bed Pressure Drop at Various Superficial Gas Velocity

particle size on axial mixing was more pronounced than that of lateral gas mixing (*Fig. 9*). In contrast to lateral gas mixing, axial gas mixing decreased with the addition of inert particles. This may be due to the small velocity gradient between bubble and dense phases with small bubble size. Therefore, convective mass transfer is reduced considerably with gas velocity in comparison with that of without inert particles. Since bubble rising velocity is proportional to $d_B^{1/2}$ bubble size would be closely related to velocity gradient and convective mass transfer. Moreover, breakage of bubble makes more uniform bubble concentration lateral and radial directions which may reduce bubble concentration in axial coordinate.

For axial gas mixing experiments, gas flow rate range was 2-5 times of $V_{mf}$ for each particle size which was considerably lower than that in lateral gas mixing condition. In this lower gas velocity range, effect of particle size on axial gas mixing was not significant since bubble size would be comparatively smaller at lower gas velocities (*Fig. 9*).

Pressure drop in the bed increased gradually with the addition of inert particles at the given gas velocities.

Typical pressure drop profiles can be seen in *Fig. 10* with the weight variation of inert particles. The addition of bigger but lighter particles into the bed produced solid partial mixing around $V_{mf}$ codition.

As can be seen in the figure that $V_{mf}$ increased exponentially with increasing weight percent of inert particles in the bed of smaller particles. The addition of inert particles in the bed reduced axial gas mixing and it improved lateral gas mixing. These may be due to the longer residence time of gas phase in the bed with inert particles.

### 3. Gas Mixing Correlation

The lateral and axial gas mixing data in the annular fluidized bed were cerrelated in terms of gas velocity and particle size since these variables are the main cause of gas mixing in fluidized beds. The linear relationship has been found between logarithmic values of axial dispersion coefficient and gas velocity.[6] Similar trend was also observed in the present study.

The Arrhenius type equation has been suggested by Baerns et al.[18] for gas mixing coefficient.

$$D_\theta \text{ or } D_z = A\exp(-B/V_g) \qquad (8)$$

in which $A$ and $B$ are the empirical constants and activation energy of particle movement, respectively,

Since $A$ and $B$ are influenced by particle size which may be written as

$$D_\theta \text{ or } D_z = A\ d_p^{n_1}\exp(-B\ d_p^{n_2}/V_g)$$

Within the present experimental conditions, gas mixing coefficients can be expressed as:

For lateral gas mixing

$$D_\theta = 93\ d_p^{0.27} \exp(-602 d_p^{0.51}/V_g) \quad (10)$$

Standard error of estimate : 0.11

For axial gas mixing

$$D_z = 1.3 \times 10^9\ d_p^{3.40} \exp(-8.93 \times 10^4 \cdot d_p^{2.02}/V_g) \quad (11)$$

Standard error of estimate : 0.13

## Conclusions

1. The addition of inert particles in the bed of small particle size enhanced lateral gas mixing and it reduced axial gas mixing.
2. Axial and lateral gas mixing coefficients increased exponentially with gas velocity.
3. Gas mixing coefficients were well represented in terms of gas flow rate and particle size in an annular fluidized bed.

## Nomenclature

$A$ : empirical constant

$B$ : empirical constant

$C$ : constant

$C_A$ : tracer concentration, g-mole/cm$^3$

$C_{Ai}$ : initial tracer concentration, g-mole/cm$^3$

$C_{A0}$ : tracer concentration after completely mixed, g-mole/cm$^3$

$D_i$ : inner bed diameter, cm

$D_0$ : outer bed diameter, cm

$d_B$ : bubble diameter, cm

$d_p$ : particle diameter, cm

$D_\theta$ : lateral gas mixing coefficient, cm$^2$/sec

$D_z$ : axial gas mixing coefficient, cm$^2$/sec

$L$ : length of fluidized bed, cm

$N$ : constant

$n_1$ : empirical constant

$n_2$ : empirical constant

$\Delta P$ : pressure drop in the bed, atm.

$\Delta P_{v\to\infty}$ : ultimate pressure drop at infinite gas velocity, atm.

$Pe$ : Peclet number based on particle diameter, $V_g\ d_p/D_z$

$t$ : time, sec

$V_g$ : gas superficial velocity, cm/sec

$V_{mf}$ : minimum fluidization velocity, cm/sec

$Z$ : axial direction coordinate

$\theta$ : lateral direction coordinate

$\sigma$ : second moment of tracer response curve about the mean

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